

## PATENT ABSTRACTS OF JAPAN

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(54) POLYMER SOLID ELECTROLYTIC AND BATTERY

(57)Abstract:

PROBLEM TO BE SOLVED: To provide polymer solid electrolyte and a battery having superior workability, moldability, mechanical strength, flexibility, and heat resistance and having a superior ion conductivity.

SOLUTION: This polymer solid electrolyte includes polyether polymer having siloxane bonding in its side-chain, electrolyte salt compound, namely presenting aprotic organic solvent, and plasticizer such as polyalkylene glycol of 200-5000 in number average molecular weight. This battery comprises the polymer solid electrolyte, a positive electrode, and a negative electrode.

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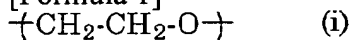
## CLAIMS

[Claim(s)]

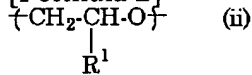
[Claim 1] The polyether polymer which has siloxane association in a side chain, and the solid polymer electrolyte characterized by including an electrolyte salt compound.

[Claim 2] or the polyether polymer which has siloxane association in a side chain is the copolymer which has the configuration unit expressed with a formula (i), the configuration unit expressed with a formula (ii), and the configuration unit expressed by the formula (iii) -- or a configuration unit (i), and (ii) -- and (iii) -- in addition, the solid polymer electrolyte according to claim 1 characterized by to be the copolymer which has further the configuration unit which be expressed with a formula (iv), and for which a bridge can be constructed

[Formula 1]

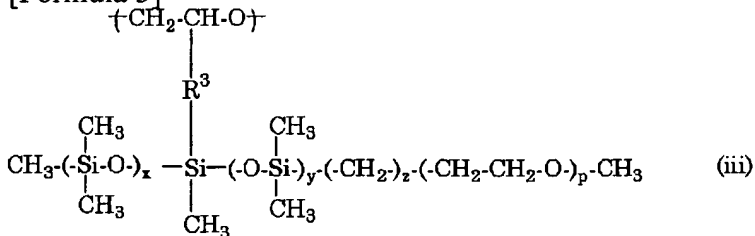


[Formula 2]

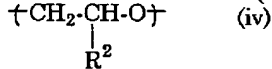


[-- R<sup>1</sup> expresses a with a carbon number of four or less alkyl group, -CH<sub>2</sub>-O-(-CH<sub>2</sub>-CH<sub>2</sub>-O-) n-CH<sub>3</sub>, or -CH<sub>2</sub>-O-CH[(-CH<sub>2</sub>-CH<sub>2</sub>-O-) n-CH<sub>3</sub>] 2 among a formula, and n shows the integer of 0-12.]

[Formula 3]



[-- the inside of a formula, and R<sup>3</sup> -- the alkylene group of carbon numbers 1-5, or (Si)-CH<sub>2</sub> -- as for 0-50z, a -CH<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>-(C) radical and x show the integer of 0-50 for 0-50y, as for 0-5p.] [Formula 4]



[-- R<sup>2</sup> shows among a formula the functional group which has a reactant radical.]

[Claim 3] The solid polymer electrolyte according to claim 2 whose R<sup>2</sup> set in the configuration unit which is expressed with a formula (iv), and for which a bridge can be constructed is (a) reactivity silicon radical, (b) methyl epoxy group, (c) ethylene nature partial saturation radical, or (d) halogen atom.

[Claim 4] The solid polymer electrolyte according to claim 1 or 2 whose weight average molecular weight of the polyether polymer which has siloxane association in a side chain is 104-107.

[Claim 5] An electrolyte salt compound is a solid polymer electrolyte according to claim 1 to 4 characterized by being a lithium salt compound.

[Claim 6] An aprotic organic solvent and the solid polymer electrolyte according to claim 1 to 5 with which number average molecular weight also contains further the plasticizer chosen from the derivative of a polyethylene glycol the straight chain mold of 200-5000, or branch-type.

[Claim 7] The solid polymer electrolyte according to claim 1 to 6 which is the crosslinked polymer over which the polyether polymer constructed the bridge using the reactant functional group.

[Claim 8] The cell which consists of a solid polymer electrolyte, a positive electrode, and a negative electrode according to claim 1 to 7.

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[Translation done.]

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DETAILED DESCRIPTION

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to a solid polymer electrolyte suitable as charges of electrochemistry device material, such as a cell, a capacitor, and a sensor, especially about a giant-molecule solid electrolyte. [0002]

[Description of the Prior Art] Conventionally, although the thing of the shape of a solution or a paste is used from the ion conductivity point, since the electrolyte which constitutes electrochemistry devices, such as a cell, a capacitor, and a sensor, needs that there is fear of damage on the device by the liquid spill, and the separator into which the electrolytic solution is infiltrated, troubles, like a limitation is in a microminiaturization of a device and thin shape-ization are pointed out. On the other hand, solid electrolytes, such as inorganic crystal nature matter, inorganic glass, and organic macromolecule system matter, are proposed. Generally the organic macromolecule system matter is excellent in workability and a moldability, the solid electrolyte obtained has flexibility and bending workability, and the progress is expected from points, like the degree of freedom of a design of the device applied becomes high. However, in an ion conductivity field, the present condition is that it is inferior to other quality of the materials.

[0003] From the ion conductivity discovery in the homopolymer of ethylene oxide, and an ant potash metal ion system, research of a solid polymer electrolyte came to be done actively. Consequently, as a polymer matrix, polyethers, such as polyethylene oxide, are considered to be the most promising in respect of the solubility of the motile height and a metal cation. It is predicted that migration of ion takes place in not the crystal section but the amorphous part of a polymer. Since then, in order to reduce the crystallinity of polyethylene oxide, copolymerization with various epoxide has been performed. The solid electrolyte which consists of a copolymer of ethylene oxide and propylene oxide, and the solid electrolyte which becomes an United States patent USP No. 4,818,644 official report from the copolymer of ethylene oxide and methyl glycidyl ether are shown at JP,62-249361,B. However, as for ionic conductivity, neither was necessarily satisfactory.

[0004] Moreover, although the attempt which a diethylene-glycol methyl glycidyl ether-ethylene oxide copolymer is made to contain a specific alkali-metal salt, and is applied to a solid polymer electrolyte is proposed by JP,9-324114,A, the value of sufficient ionic conductivity is not acquired practical.

[0005]

[Problem(s) to be Solved by the Invention] The purpose of this invention is to offer the solid polymer electrolyte which is excellent in workability, a moldability, a mechanical strength, flexibility, thermal resistance, etc., and is excellent also in ion conductivity.

[0006]

[Means for Solving the Problem] This inventions are the polyether polymer which has siloxane association in a side chain, and a solid polymer electrolyte characterized by including an electrolyte salt compound. (C) (B) electrolyte salt compound and C [ the polyether polymer which has siloxane association in the (A) side chain in detail, and ] If it requires, they will be the existing aprotic organic solvent and the solid polymer electrolyte characterized by number average molecular weight containing the plasticizer chosen from the derivative of a polyalkylene glycol the straight chain mold of 200-5000, or branch-type.

[0007] A polyether polymer (A) is a polymer which may have siloxane association (-SiO-) in a side chain, and may have the reactant functional group in which crosslinking reaction is still more possible.

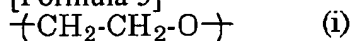
[0008] The manufacture approach of a polyether polymer for which a bridge can be constructed of having

siloxane association in a side chain has the approach of carrying out copolymerization with the oxirane compound which has siloxane association, or the approach of carrying out the graft of the polysiloxane compound which has a Si-H radical to the polyether polymer which has a partial saturation radical by the hydrosilylation reaction.

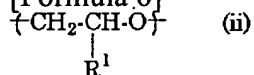
[0009] By the approach of manufacturing by copolymerization with the oxirane compound which has siloxane association, a polyether polymer can be obtained by copolymerizing the oxirane compound (ethylene oxide) which guides (I) configuration unit (i), the oxirane compound which guides (II) configuration unit (ii), and the oxirane compound which guides a configuration (III) unit (iii). In addition to an oxirane compound (I) - (III), the oxirane compound which has the reactant radical which guides (IV) configuration unit (iv), and for which a bridge can be constructed may be copolymerized further.

[0010]

[Formula 5]



[Formula 6]

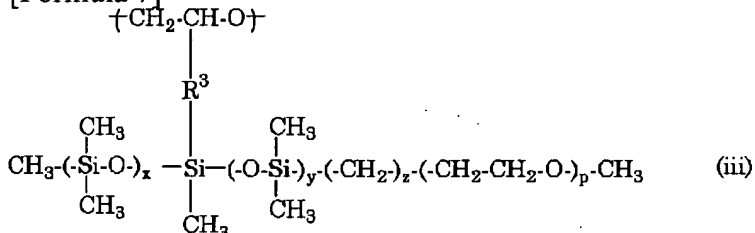


[-- R<sup>1</sup> expresses a with a carbon number of four or less alkyl group, -CH<sub>2</sub>-O-(-CH<sub>2</sub>-CH<sub>2</sub>-O-) n-CH<sub>3</sub>, or -CH<sub>2</sub>-O-CH[(-CH<sub>2</sub>-CH<sub>2</sub>-O-) n-CH<sub>3</sub>] 2 among a formula, and n shows the integer of 0-12.]

In a formula (ii), the examples of R<sup>1</sup> may be a methyl group, an ethyl group, a propyl group, and butyl, and n may be the integer of 0-8, especially 1-4.

[0011]

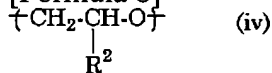
[Formula 7]



[-- the inside of a formula, and R<sup>3</sup> -- the alkylene group of carbon numbers 1-5, or (Si)-CH<sub>2</sub> -- as for 0-50z, a -CH<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>-(C) radical and x show the integer of 0-50 for 0-50y, as for 0-5p.] [0012] a formula (iii) -- setting -- the example of R<sup>3</sup> -- a methyl group, an ethyl group, a propyl group, butyl, and a pentyl radical -- it is -- x and y -- 0-50 -- especially -- the integer of 0-20 -- it is -- z -- 0-5 -- especially -- the integer of 0-3 -- it is -- p -- 0-50 -- it is the integer of 0-30 especially. [ for example, ] [ for example, ] [ for example, ]

[0013]

[Formula 8]

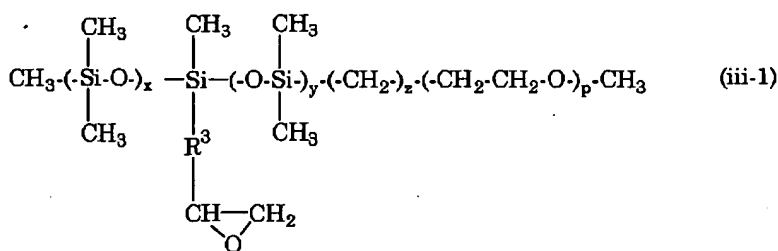


[-- R<sup>2</sup> shows among a formula the functional group which has a reactant radical.]

[0014] The oxirane compound (I) which guides a configuration unit (i) is ethylene oxide. Alkylene oxide, a glycidyl ether compound, etc. which may have the substituent are mentioned to the oxirane compound (II) which guides a configuration unit (ii). Specifically, oxirane compounds, such as propylene oxide, methyl glycidyl ether, butyl-glycidyl-ether, styrene oxide, phenyl-glycidyl-ether, 1, 2-epoxy hexane, ethylene glycol methyl glycidyl ether, diethylene-glycol methyl glycidyl ether, triethylene glycol methyl glycidyl ether, 1, 3-bis (2-methoxyethoxy) propane 2-glycidyl ether, 1, and 3-screw [2-(2-methoxyethoxy) ethoxy] propane 2-glycidyl ether, are mentioned.

[0015] The example of the oxirane compound (III) which has siloxane association which guides a configuration unit (iii) is a compound expressed with a formula (iii-1).

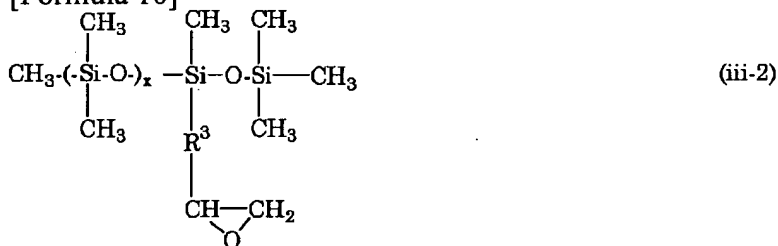
[Formula 9]



[-- the inside of a formula, and R<sup>3</sup> -- the alkylene group of carbon numbers 1-5 -- or -- -- (Si)-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>- as for 0-50z, (C) and x show the integer of 0-50 for 0-50y, as for 0-5p.]

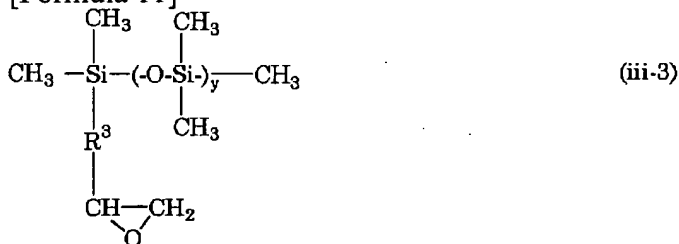
[0016] An oxirane compound (iii-1) is a compound preferably expressed with a formula (iii-2) and a formula (iii-3).

[Formula 10]



[-- the inside of a formula, and R<sup>3</sup> -- the alkylene group of carbon numbers 1-5 -- or -- -- (Si)-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>- (C) and x show the integer of 1-10.]

[Formula 11]



[-- the inside of a formula, and R<sup>3</sup> -- the alkylene group of carbon numbers 1-5 -- or -- -- (Si)-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>- (C) and y show the integer of 0-10.]

[0017] The example of the reactant functional group of the oxirane compound which has the reactant functional group which guides a configuration unit (iv) is (a) reactivity silicon radical, (b) methyl epoxy group, (c) ethylene nature partial saturation radical, or (d) halogen atom.

[0018] (a) To the oxirane compound which has a reactant silicon radical 2-glycidoxy ethyl trimethoxy silane, 3-glycidoxypropylmethyldimethoxysilane, 3-glycidoxypropyltrimethoxysilane, 4-glycidoxybutyl methyl trimethoxysilane, 3-(1, 2-epoxy) propyltrimethoxysilane, 4-(1, 2-epoxy) butyltrimethoxysilane, 5-(1, 2-epoxy) pentyl trimethoxysilane, 1-(3, 4-epoxycyclohexyl) methyl methyl dimethoxysilane, and 2-(3, 4-epoxycyclohexyl) ethyltrimethoxysilane are mentioned. In these, 3-glycidoxypropyltrimethoxysilane and especially 3-glycidoxypropylmethyldimethoxysilane are desirable.

[0019] (b) To the oxirane compound which has a methyl epoxy group 2, 3-epoxy propyl - 2', the 3' - epoxy -2'-methylpropyl ether, Ethylene GUKORU -2, 3-epoxy propyl - 2', the 3' - epoxy -2'-methylpropyl ether, And a diethylene glycol -2, 3-epoxy propyl - 2', the 3' - epoxy -2'-methylpropyl ether, 2-methyl - 1, 2, 3, 4-diepoxy butane, 2-methyl - 1, 2, 4, 5-diepoxy pentane, 2-methyl - 1, 2, 5, 6-diepoxy hexane, a hydroquinone -2, 3-epoxy propyl - 2', the 3' - epoxy -2'-methylpropyl ether, A catechol -2, 3-epoxy propyl - 2', the 3' - epoxy -2'-methylpropyl ether, etc. are mentioned.

[0020] the inside of it -- especially -- 2 and 3-epoxy propyl-2' and 3' - epoxy -2 -- ' - methylpropyl ether and ethylene glycol -2, 3-epoxy propyl -2', and the 3' - epoxy -2'-methylpropyl ether are desirable.

[0021] (c) To the oxirane compound which has an ethylene nature partial saturation radical Allyl glycidyl ether, 4-vinyl cyclohexyl glycidyl ether, alpha-TERUPI nil glycidyl ether, cyclohexenyl methyl glycidyl ether, p-

vinylbenzyl glycidyl ether, allyl compound phenyl glycidyl ether, Vinyl glycidyl ether, 3, 4-epoxy-1-butene, 3, 4-epoxy-1-pentene, 4, a 5-epoxy-2-pentene, 1, 2-epoxy -5, 9-cyclo dodeca diene, 3, 4-epoxy-1-vinyl cyclohexene, 1, 2-epoxy-5-cyclooctane, metaglycidyl acrylate, glycidyl methacrylate, sorbic-acid glycidyl, cinnamic-acid glycidyl, crotonic-acid glycidyl, and glycidyl-4-HEKISENOETO is used. Preferably, allyl glycidyl ether, metaglycidyl acrylate, and glycidyl methacrylate are mentioned.

[0022] (d) Epibromohydrin, EPIYODOHIDORIN, and epichlorohydrin are mentioned to the oxirane compound which has a halogen atom.

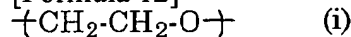
[0023] The polymerization method of a polyether polymer for having siloxane association in a side chain is the polymerization method for obtaining a polymer by the ring opening reaction of an ethyleneoxide part, and is performed to these people's JP,63-154736,A and JP,62-169823,A like the approach of a publication.

[0024] A degree requires a polymerization reaction and it can be performed. A polyether polymer is obtained using the catalyst system which makes organic aluminium a subject as a catalyst for ring opening polymerization, the catalyst system which makes organic zinc a subject, an organic tin-phosphoric ester condensate catalyst system, etc. by making each monomer react under the reaction temperature of 10-80 degrees C, and churning under existence of a solvent or un-existing. Especially, points, such as polymerization degree of the polymer made and a property, to especially an organic tin-phosphoric ester condensate catalyst system is desirable. In a polymerization reaction, a reactant functional group does not react but the polyether polymer which has a reactant functional group is obtained.

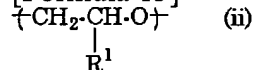
[0025] The graft of the polysiloxane compound which has a Si-H radical is carried out to the polyether polymer which has a partial saturation radical by the hydrosilylation reaction. By the approach of obtaining the polyether polymer which has siloxane association in a side chain The oxirane compound (I) which guides a configuration unit (i), and (ethylene oxide), With the oxirane compound (IV) which has the reactant radical (partial saturation radical) which guides a configuration unit (iv) for the oxirane compound (II) which guides a configuration unit (ii) as occasion demands, and for which a bridge can be constructed A polyether polymer is obtained by carrying out the graft of the polysiloxane compound which has the Si-H radical shown by the formula (v) to the polymer obtained by copolymerizing by the hydrosilylation reaction.

[0026]

[Formula 12]



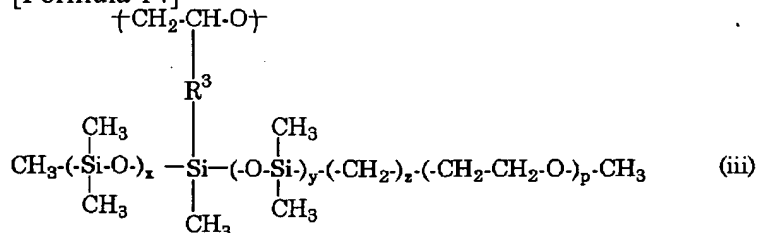
[Formula 13]



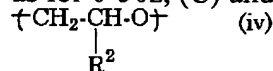
[-- R1 expresses a with a carbon number of four or less alkyl group, -CH<sub>2</sub>-O-(-CH<sub>2</sub>-CH<sub>2</sub>-O-) n-CH<sub>3</sub>, or -CH<sub>2</sub>-O-CH[(-CH<sub>2</sub>-CH<sub>2</sub>-O-) n-CH<sub>3</sub>] 2 among a formula, and n shows the integer of 0-12.]

[0027]

[Formula 14]



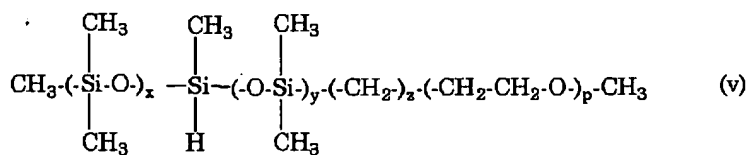
[-- the inside of a formula, and R3 -- the alkylene group of carbon numbers 1-5, or (Si)-CH<sub>2</sub>-CH<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>- as for 0-50z, (C) and x show the integer of 0-50 for 0-50y, as for 0-5p.] [Formula 15]



[-- R2 shows among a formula the functional group which has a reactant radical.]

[Formula 16]





[-- as for 0-50y, 0-5p show the integer of 0-50 for x among a formula, as for 0-50z.]

[0028] As an example of the catalyst of a hydrosilylation reaction, transition metals, such as palladium and platinum, or those compounds, and a complex are mentioned. Moreover, a peroxide, an amine, and a phosphine are also used. As for the most general catalyst, platinum complexes, such as dichlorobis (acetonitrile) palladium (II), a chloro tris (triphenyl phosphine) rhodium (I), chloroplatinic acid, and a platinum carbonyl vinyl methyl complex, are mentioned.

[0029] For the rate of the configuration unit (i) included in the polyether polymer used for the solid polymer electrolyte of this invention, the rate of a configuration unit (ii) is [ the rate of a configuration-two to 60% of the weight unit (iv) of the rate of a configuration unit (iii) ] 0 - 20 % of the weight zero to 30% of the weight two to 70% of the weight ten to 70% of the weight ten to 80% of the weight 20 to 80% of the weight preferably ten to 80% of the weight.

[0030] Ion conductivity is more high in the rate of a configuration unit (iii) being 2 % of the weight or more. Moreover, the solubility of an electrolyte salt compound is good in it being 90 or less % of the weight, and ion conductivity is high.

[0031] although it was known that ion conductivity will improve by generally lowering glass transition temperature, in the case of the polyether polymer of this invention, it was markedly alike, and it found that the ion conductivity improvement effectiveness was large.

[0032] In order to acquire good workability, a moldability, a mechanical strength, and flexibility, the thing of 105 to 5x10<sup>6</sup> within the limits of the molecular weight of a polyether polymer is preferably desirable within the limits of weight average molecular weight 104-107.

[0033] In this invention, -65 degrees C or less and the amount of heat of fusions have [ the glass transition temperature of a polyether polymer ] a desirable thing 60J [g ] or less. Ion conductivity is high when glass transition temperature and the amount of heat of fusions are in the above-mentioned value. The glass transition temperature and the amount of heat of fusions of a polyether polymer are measured with a differential scanning calorimeter (DSC).

[0034] As for the polyether polymer of this invention, it is desirable that it is a copolymer. Which type of a block copolymer and a random copolymer is sufficient as a polyether polymer. Since the effectiveness that the random copolymer reduces the crystallinity of polyethylene oxide more is large, it is desirable.

[0035] As the bridge formation approach of the polymer which is a reactant silicon radical, a reactant functional group can construct a bridge by the reaction of a reactant silicon radical and water. In order to raise reactivity, amine system compounds, such as organometallic compounds, such as aluminium compounds, such as aluminum, such as titanium compounds, such as tin compounds, such as dibutyltin dilaurate and dibutyltin malate, tetrabutyl titanate, and tetra-propyl titanate, aluminum tris acetylacetonato, and aluminum tris ethyl acetoacetate, or a butylamine, and an octyl amine, etc. may be used as a catalyst.

[0036] Polyamine and acid anhydrides are used in the bridge formation approach of a polyether polymer that a reactant functional group is a methyl epoxy group. As polyamine, aromatic series polyamine, such as aliphatic series polyamine [ , such as diethylenetriamine and dipropylenetriamine, ], 4, and 4'-diamino diphenyl ether, diaminodiphenyl sulfone, m-phenylenediamine, and xylylene diamine, etc. is mentioned. Although the addition of polyamine changes with classes of polyamine, it is usually 0.1 - 10% of the weight of the range of the whole (namely, constituent excluding the plasticizer from the solid electrolyte) constituent except a plasticizer.

[0037] As acid anhydrides, a maleic anhydride, phthalic anhydride, methyl hexahydro phthalic anhydride, a tetramethylen maleic anhydride, tetrahydro phthalic anhydride, etc. are mentioned. Although the addition of acid anhydrides changes with classes of acid anhydride, it is usually 0.1 - 10% of the weight of the range of the whole constituent except a plasticizer. An accelerator may be used for these bridge formation, there are a phenol, cresol, resorcinol, etc. in the crosslinking reaction of polyamine, and there are benzyl dimethylamine, 2-(dimethylaminoethyl) phenol, dimethylaniline, etc. in the crosslinking reaction of acid anhydrides. Although the addition of an accelerator changes with accelerators, it is usually 0.1 - 10% of the weight of the range of a cross

linking agent. [0038] As the bridge formation approach of a polyether polymer that a reactant functional group is an ethylene nature partial saturation radical, activity energy lines, such as a radical initiator chosen from organic peroxide, an azo compound, etc., ultraviolet rays, and an electron ray, are used. Furthermore, the cross linking agent which has silicon hydride can also be used.

[0039] As organic peroxide, what is usually used for the bridge formation application is used, and ketone peroxide, peroxy ketal, hydroperoxide, dialkyl peroxide, diacyl peroxide, peroxy ester, etc. are 1 and 1-screw (tert-butyl peroxide). - 3, 3, a 5-trimethyl cyclohexane, di-t-butyl peroxide, t-butyl cumyl peroxide, dicumyl peroxide, 2, the 5-dimethyl -2, 5-di-tert-butyl peroxide hexane, benzoyl peroxide, etc. are mentioned. Although the addition of organic peroxide changes with classes of organic peroxide, it is usually within the limits of 0.1 - 10% of the weight of the whole constituent except a plasticizer.

[0040] As an azo compound, an azonitrile compound, an azo amide compound, an azo amidine compound, etc., What is used for the bridge formation application is used. Usually, 2 and 2'-azobisisobutyronitril, - azobis (2-methyl butyronitrile), and 2 and 2' 2, 2'-azobis (4-methoxy-2,4-dimethylvaleronitrile), 2 and 2'-azobis (2-methyl-N-phenyl propione amidine) dihydrochloride, 2 and 2'-azobis [2-(2-imidazoline-2-IRU) propane], - azobis (isobutane), and 2 and 2'-azobis [2-methyl-N-(2-hydroxyethyl) propione amide], 2, and 2' 2, 2'-azobis [2-(hydroxymethyl) propionitrile] etc. is mentioned. Although the addition of an azo compound changes with classes of azo compound, it is usually within the limits of 0.1 - 10% of the weight of the whole constituent except a plasticizer.

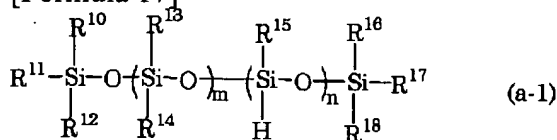
[0041] Especially in bridge formation by the activity energy-line exposure of ultraviolet rays etc., allyl glycidyl ether, metaglycidyl acrylate, glycidyl methacrylate, and cinnamic-acid glycidyl are desirable among reactant functional-group content monomer components. Moreover, it is a diethoxy acetophenone and 2-hydroxy as a sensitization assistant. - 2-methyl-1-phenyl propane-1-ON, Benzoin ether, such as acetophenones, such as phenyl ketone, a benzoin, and benzoin methyl ether, Benzophenones, such as a benzophenone and 4-phenylbenzo phenon Azides, such as thioxan tons, such as 2-isopropyl thioxan ton, 2, and 4-dimethyl thioxan ton, 3-sulfonyl azide benzoic acid, and 4-sulfonyl azide benzoic acid, can be used for arbitration.

[0042] Ethylene glycol diacrylate, ethylene glycol dimethacrylate, oligo ethylene glycol diacrylate, oligo ethylene glycol dimethacrylate, an allyl compound METARI crate, allyl compound acrylate, diallyl malete, triallyl isocyanurate, bis-phenyl maleimide, a maleic anhydride, etc. can be used for arbitration as a bridge formation assistant.

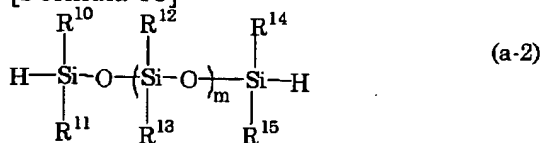
[0043] As a compound which has the silicon hydride which constructs a bridge in an ethylene nature partial saturation radical, the compound which has at least two silicon hydrides is used. A polysiloxane compound or a polysilane compound is especially good. the line expressed with a formula (a-1) or a formula (a-2) as a polysiloxane compound -- there is a polysiloxane compound or an annular polysiloxane compound expressed with a formula (a-3).

[0044]

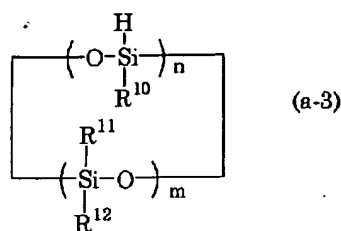
[Formula 17]



[Formula 18]



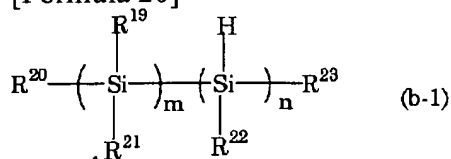
[Formula 19]



[0045] however, a formula (a-1) - formula (a-3) type -- setting -- R10, R11, R12, R13, R14, R15 and R16, and R17 and -- R18 expresses the alkyl group or alkoxy group of a hydrogen atom or carbon numbers 1-12, and expresses  $n \geq 2$ ,  $m \geq 0$ , and  $2 \leq m+n \leq 300$ . As an alkyl group, low-grade alkyl groups, such as a methyl group and an ethyl group, are desirable. As an alkoxy group, lower alkoxy groups, such as a methoxy group and an ethoxy radical, are desirable.

[0046] the line expressed with a formula (b-1) as a polysilane compound -- a polysilane compound is used.

[Formula 20]



However, it sets at a ceremony (b-1), and is R19, R20, R21, and R22 and R23. The alkyl group or alkoxy group of a hydrogen atom or carbon numbers 1-12 is expressed, and  $n \geq 2$ ,  $m \geq 0$ , and  $2 \leq m+n \leq 100$  are expressed.

[0047] As an example of the catalyst of a hydrosilylation reaction, transition metals, such as palladium and platinum, or those compounds, and a complex are mentioned. Moreover, a peroxide, an amine, and a phosphine are also used. As for the most general catalyst, dichlorobis (acetonitrile) palladium (II), a chloro tris (triphenyl phosphine) rhodium (I), and chloroplatinic acid are mentioned.

[0048] As the bridge formation approach of the polyether polymer halogen atom content, cross linking agents, such as polyamine, mercapto imidazoline, mercaptopyrimidine, thiourea, and the Pori mercaptans, are used. Triethylenetetramine, a hexamethylenediamine, etc. are mentioned as polyamine. As mercapto imidazoline, 2-mercapto imidazoline, 4-methyl-2-mercapto imidazoline, etc. are mentioned. As mercaptopyrimidine, 2-mercapto pyrimidine, 4, and 6-dimethyl-2-mercapto pyrimidine etc. is mentioned. Ethylene thiourea, dibutyl thiourea, etc. are mentioned as thiourea. As Pori mercaptans, 2-dibutylamino -4, 6-dimethyl KAPUTO-s-triazine, 2-phenylamino -4, 6-dimercapto triazine, etc. are mentioned. Although the addition of a cross linking agent changes with classes of cross linking agent, it is usually 0.1 - 30% of the weight of the range of the whole constituent except a plasticizer.

[0049] Moreover, it is effective from the standpoint of the thermal stability of a halogen content polymer to add the metallic compounds which serve as carrier acid further to a solid polymer electrolyte. as the metallic oxide used as such carrier acid -- the [ periodic-table ] -- there are the oxide of the oxide of II group metal, a hydroxide, a carbonate, carboxylate, a silicate, a borate, phosphite, and a periodic-table VIa group metal, a basic carbonate, basic carboxylate, basic phosphite, a basic sulfite, a 3 basicity sulfate, etc. As a concrete example, a magnesia, a magnesium hydroxide, a magnesium carbonate, a calcium silicate, calcium stearate, a minium, tin stearate, etc. can be mentioned. Although the loadings of the metallic compounds used as the above-mentioned carrier acid change with classes; it is usually 0.1 - 30% of the weight of the range of the whole constituent except a plasticizer.

[0050] or [ that a bridge is not constructed over the electrolyte salt compound used in this invention ] -- or it is desirable that it is meltable into the polyether polymer over which the bridge is constructed, and the mixture which consists of a plasticizer. In this invention, the next salt compound is used preferably.

[0051] Namely, a metal cation, ammonium ion, friend JINIUMU ion, and the cation chosen from GUANIJIUMU ion, A chlorine ion, bromine ion, iodine ion, perchloric acid ion, thiocyanic acid ion, Tetrafluoro boron acid ion, nitrate ion, AsF6-, PF6-, Stearyl sulfonic-acid ion, octyl sulfonic-acid ion, the dodecylbenzenesulfonic acid ion, Naphthalene sulfonic-acid ion, dodecyl naphthalene sulfonic-acid ion, 7, 7, 8, and 8-tetracyano-p-quinodimethan ion, X1SO3-, The compound which consists of an anion chosen from [(X1SO2) (X2SO2) N]-, [(X1SO2) (X2SO2) (X3SO2) C]-, and [(X1SO2) (X2SO2) YC]- is mentioned.

However, X1, X2, X3, and Y are electronic suction nature machines. The carbon numbers of X1, X2, and X3 are the perfluoroalkyl radicals or perfluoro aryl groups from 1 to 6 respectively independently preferably, and Y is a nitro group, a nitroso group, a carbonyl group, a carboxyl group, or a cyano group. Even if X1, X2, and X3 are respectively the same, they may differ. The cation of transition metals can be used as a metal cation. The cation of the metal preferably chosen from Mn, Fe, Co, nickel, Cu, Zn, and Ag metal is used. Moreover, a desirable result is obtained even if it uses the cation of the metal chosen from Li, Na, K, Rb, Cs, Mg, calcium, and Ba metal. It is free to use together two or more kinds of above-mentioned compounds as an electrolyte salt compound. [0052] this invention -- setting -- the amount of the electrolyte salt compound used -- the polyether polymer 100 weight section -- receiving -- 1 - 50 weight section -- the range of 3-20-fold section is preferably good. Workability, a moldability, and the mechanical strength and flexibility of a solid electrolyte that were acquired are good in the amount of an electrolyte salt compound being below 50 weight sections, and ion conductivity is also still higher.

[0053] An aprotic organic solvent or number average molecular weight of a plasticizer is a polyalkylene glycol the straight chain mold of 200-5000, or branch-type. As an aprotic organic solvent, aprotic ether and ester are desirable. Specifically Propylene carbonate, gamma-butyrolactone, butylene carbonate, Ethylene carbonate, dimethyl carbonate, ethyl methyl carbonate, Diethyl carbonate, 1, 2-dimethoxyethane, 1, 2-JIMETOKI propane, 3-methyl-2-oxazolidone, a tetrahydrofuran, 2-methyl tetrahydrofuran, The 1, 3-dioxolane, 4, and 4-methyl -1, 3-dioxolane, tert-butyl ether, iso-butyl ether, 1, 2-ethoxy methoxyethane, diethylene-glycol wood ether, Diethylene-glycol diethylether, triethylene glycol wood ether, Triethylene glycol diethylether, tetraethylene glycol wood ether, Tetraethylene glycol diethylether, ethylene glyme, an ethylene jig lime, methyl tetraglyme, a methyl TORIGU lime, a methyl jig lime, methyl formate, methyl acetate, methyl propionate, etc. are mentioned, and two or more sorts of such mixture may be used. Especially desirable things are propylene carbonate, gamma-butyrolactone, butylene carbonate, and 3-methyl-2-oxazoline. Moreover, diethylene-glycol wood ether, diethylene-glycol diethylether, triethylene glycol wood ether, triethylene glycol diethylether, tetraethylene glycol wood ether, and tetraethylene glycol diethylether are also especially desirable organic solvents.

[0054] As a derivative of a straight chain mold or branch-type polyalkylene glycol, number average molecular weight is obtained from the polyalkylene glycol of 200-5000. A polyethylene glycol or a polypropylene glycol is mentioned as a polyalkylene glycol, and there is the ester derivative or ether derivative which consists of an alkyl group of carbon numbers 1-8 and an alkenyl radical of carbon numbers 3-8 as the derivative.

[0055] As an ether derivative, diester, such as polyalkylene GURIKORUJI acetic ester (for example, polyethylene GURIKORUJI acetic ester), can be mentioned among derivatives as diethers, such as wood ether, diethylether, dipropyl ether, and diaryl ether, and an ester derivative.

[0056] The still more desirable range of the number average molecular weight of the polyalkylene glycol to be used is 200-2000. although the blending ratio of coal of a plasticizer is arbitrary -- the polyether polymer 100 weight section -- receiving -- the 0 - 2000 weight section -- it is the 5 - 1000 weight section preferably.

[0057] In case a solid polymer electrolyte is used, a flame retarder can be used when fire retardancy is required. As a flame retarder, it chooses from halogenides, such as a bromination epoxy compound, tetra-bromine bisphenol A, and chlorinated paraffin, an antimony trioxide, antimony pentoxide, an aluminum hydroxide, a magnesium hydroxide, phosphoric ester, a polyphosphate, and boric-acid zinc, and an effective dose is added.

[0058] What is necessary is just to usually mix each component mechanically, although the manufacture approach of the solid polymer electrolyte of this invention does not have especially constraint. In the case of the polyether polymer which needs bridge formation, mechanically, after mixing, long duration immersion may be carried out and each component may be infiltrated into a plasticizer after bridge formation, although manufactured by the approach of making a bridge construct etc. As a means to mix mechanically, various kneaders, an opening roll, an extruder, etc. can be used for arbitration.

[0059] When a reactant functional group is a reactant silicon radical, especially since it happens easily also with the moisture in an ambient atmosphere, the amount of the water used for crosslinking reaction is not restricted. A bridge can also be constructed by letting it pass to short-time cold water or a warm water bath, or exposing to a steam ambient atmosphere.

[0060] If a radical initiator is used when a reactant functional group is an ethylene nature partial saturation radical, crosslinking reaction will be completed in 1 minute - bottom 20 hours of a temperature condition of 10 degrees C - 200 degrees C. Moreover, when using energy lines, such as ultraviolet rays, generally a sensitizer is

used. Usually, crosslinking reaction is completed in 0.1 seconds - bottom 1 hour of a temperature condition of 10 degrees C - 150 degrees C. Crosslinking reaction is completed at the cross linking agent which has silicon hydride in 10 minutes - bottom 10 hours of a temperature condition of 10 degrees C - 180 degrees C.

[0061] Although especially the approach of mixing an electrolyte salt compound and a plasticizer to a polyether polymer is not restrained, after melting the approach or the polyether polymer with which a plasticizer makes melt and mix the approach, the polyether polymer, and the electrolyte salt compound with which the approach, the electrolyte salt compound, and the plasticizer which long-duration immersion is carried out [ plasticizer ] and infiltrates a polyether polymer into the organic solvent containing an electrolyte salt compound and a plasticizer mix mechanically to a polyether polymer to an organic solvent besides once, there is a method of mixing a plasticizer etc. the case where it manufactures using an organic solvent -- various kinds of polar solvents, for example, a tetrahydrofuran, an acetone, an acetonitrile, dimethylformamide, dimethyl sulfoxide, dioxane, a methyl ethyl ketone, methyl isobutyl ketone, etc. -- independence -- or it is mixed and used.

[0062] The solid polymer electrolyte shown by this invention is excellent in a mechanical strength and flexibility, and considering as the solid electrolyte of a large area thin film configuration using the property is acquired easily. For example, production of the cell using the polyelectrolyte of this invention is possible. In this case, as a positive-electrode ingredient, there are a lithium-manganese multiple oxide, a cobalt acid lithium, vanadium pentoxide, olivine mold phosphoric-acid iron, polyacetylene, the poly pyrene, the poly aniline, polyphenylene, polyphenylene sulfide, polyphenylene oxide, polypyrrole, Pori Fran, a poly azulene, etc. There are the intercalation compound and lithium metal with which occlusion of the lithium was carried out between graphite or the layer of carbon as a negative-electrode ingredient, a lithium-lead alloy, etc. Moreover, the use as a diaphragm of the ion electrode of cations, such as alkali-metal ion, Cu ion, calcium ion, and Mg ion, is also considered using high electrical conductivity. Especially the giant-molecule solid electrolyte of this invention is suitable as charges of electrochemistry device material, such as a cell, a capacitor, and a sensor.

[0063]

[Example] Hereafter, an example is shown and this invention is explained concretely.

[0064] In the following examples, analysis was performed as follows. The monomer conversion presentation of polyether plural copolymers is <sup>1</sup>H NMR. It asked with the spectrum. Gel-permeation-chromatography measurement was carried out to the molecular weight measurement of polyether plural copolymers, and molecular weight was computed by standard polystyrene conversion. Gel-permeation-chromatography measurement is the measuring device of Shimadzu Corp. Shodex of RID-6A and the column by Showa Denko K.K. KD-807, KD-806, and KD-806M And KD-803 and a solvent DMF (dimethylformamide) It uses. It carried out at 60 degrees C.

[0065] Glass transition temperature and the amount of heat of fusions are a differential scanning calorimeter made from Physical science Electrical and electric equipment. DSC8230B It uses and they are the inside of nitrogen-gas-atmosphere mind, and a temperature requirement. -100-80 degrees C, programming rate 10 degrees C / min It measured. In order to measure conductivity sigma, it is a sample film beforehand. 70 degrees C and a 12-hour vacuum drying were performed. Measurement of conductivity It carries out at 15 degrees C and is a film. SUS It inserts with the electrode of make and is an electrical potential difference. 0.5V, frequency range 5Hz - 1MHz It computed by the complex impedance method using the alternating current anodizing process.

[0066] The synthetic example 1 (manufacture of a catalyst)

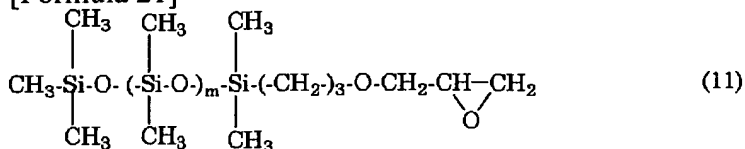
Having put tributyl tin chloride 10g and tributyl phosphate 35g into 3 opening flask equipped with the agitator, the thermometer, and the distillation apparatus, and agitating under a nitrogen air current, heated for 20 minutes at 250 degrees C, the distillate was made to distill off, and solid-state-like the quality of a condensate was obtained as the residue. This was used as a catalyst for polymerizations after that.

[0067] Example 1 inner capacity 3L The nitrogen purge of the interior of a four glass opening flask is carried out. Propylene oxide 20g adjusted to the quality of a condensate of 5g shown in this in the example of manufacture of a catalyst as a catalyst, and the moisture of 10 ppm or less, Teaching n-hexane 2,000g as allyl-glycidyl-ether 8g and a solvent, and pursuing the conversion of allyl glycidyl ether with a gas chromatography ethylene oxide 124g It adds serially and the conversion of allyl glycidyl ether 200g (formula (11)) of oxirane compounds which have a polysiloxane 20% It sometimes added. The polymerization reaction stopped with the methanol. In the bottom ordinary temperature of reduced pressure after taking out a polymer by the decantation

It is under reduced pressure further for 24 hours. At 45 degrees C It dries for 10 hours and is a polymer. 150g was obtained. The glass transition temperature of this copolymer was [ 850,000 and the amount of heat of fusions of -72 degrees C and weight average molecular weight ] 34 J/g.

[0068]  $^1\text{H}$  NMR 9 % of the weight (formula (11)) of oxirane compounds with which the monomer conversion component-analysis result of this polymer by the spectrum has a polysiloxane, and ethylene oxide 76 % of the weight, 11 % of the weight of propylene oxide, and allyl glycidyl ether 4 % of the weight it was .

[Formula 21]

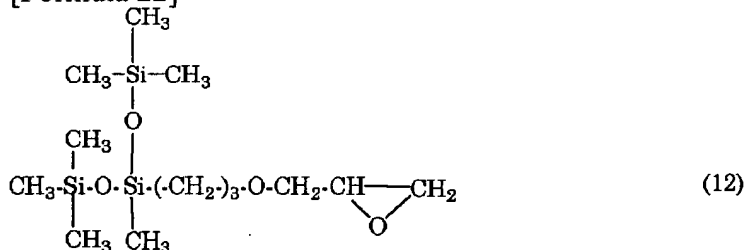


(Mn 2,000)

[0069] Glass [ of example 2 inner-capacity 3L ] The nitrogen purge of the interior of 4 opening flask is carried out. Diethylene-glycol methyl glycidyl ether 160g adjusted to the quality of a condensate of 5g shown in this in the example of manufacture of a catalyst as a catalyst, and the moisture of 10 ppm or less, Teaching n-hexane 2,000g as 3 and 4-epoxy-1 butene ether 15g and a solvent, and pursuing the conversion of the 3 and 4-epoxy-1 butene ether with a gas chromatography ethylene oxide 340g It added serially and the conversion of the 3 and 4-epoxy-1 butene ether added at o'clock 320g (formula (12)) of oxirane compounds which have siloxane association 20%. The polymerization reaction stopped with the methanol. After taking out a polymer by the decantation, it dried at 45 degrees C under reduced pressure further by 40 degrees C under ordinary pressure for 10 hours for 24 hours, and polymer 542g was obtained. The glass transition temperature of this copolymer was [ 1,050,000 and the amount of heat of fusions of -76 degrees C and weight average molecular weight ] 3 J/g.

[0070]  $^1\text{H}$  NMR 18 % of the weight (formula (12)) of oxirane compounds with which the monomer conversion component-analysis result of this polymer by the spectrum has siloxane association, and diethylene-glycol methyl glycidyl ether 24 % of the weight and ethylene oxide 56 % of the weight and the 3 and 4-epoxy-1 butene ether 2 % of the weight it was .

[Formula 22]



[0071] It is acetonitrile 20ml about 1g [ of polyether copolymers obtained in the example 3 example 1 ], and organic peroxide dicumyl peroxide 0.015g. It dissolves and is LiTFSI0.15(lithium bis(trifluoromethylsulfonyl) imide) g. 5ml of tetrahydrofuran solutions was mixed. After carrying out the cast of this mixed liquor on the mold made from polytetrafluoroethylene and drying, under nitrogen-gas-atmosphere mind, it heated for 30 minutes and 140 degrees C of bridge formation films were obtained. The conductivity of a solid electrolyte was  $2.5 \times 10^{-4}$  S/cm.

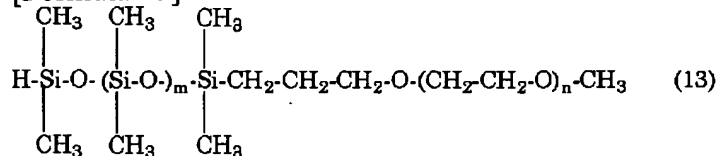
[0072] Polyether copolymer obtained in the example 4 example 2 It is an acetonitrile about 1g and organic peroxide benzoyl oxide 0.015g. 20ml It dissolves and is LiTFSI 0.15g. 5ml of tetrahydrofuran solutions was mixed. After carrying out the cast of this mixed liquor on the mold made from polytetrafluoroethylene and drying, under nitrogen-gas-atmosphere mind, 3hr heating was carried out and 100 degrees C of bridge formation films were obtained. The conductivity of a solid electrolyte was  $4.2 \times 10^{-4}$  S/cm.

[0073] The nitrogen purge of the interior of the glass flask of 5300ml of examples is carried out, and it is a methyl ethyl ketone. 100ml It adds. Ethylene oxide / triethylene glycol methyl glycidyl ether / allyl-glycidyl-ether = 47/47/6 3 yuan copolymer of weight % (weight average molecular weight is 1,050,000) Ten g, Chloroplatinic acid 0.3g, LiTFSI 0.15g, and Si-H They are both ends as 1g (formula (13)) of polysiloxane compounds which it has, and a cross linking agent. Si-H 1g (formula (14)) of polysiloxane compounds which it

has It was made to add and dissolve. This mixed liquor 10ml The cast was carried out on the mold made from polytetrafluoroethylene, the solvent was removed, and the bridge formation film was obtained 1 hour after at 40 degrees C. The property of a bridge formation film was measured by the same approach as an example 1. conductivity of a solid electrolyte  $2.3 \times 10^{-4}$  S/cm it was .

[0074]

[Formula 23]



(n/m=2, Mn 1,500)

[Formula 24]



[(Mn 500) 0075] The polymerization was performed by the same actuation as an example 1, without using the oxirane compound which has example of comparison 1 siloxane association. The monomer conversion component-analysis results of the obtained copolymer were 80 % of the weight of ethylene oxide, 16 % of the weight of propylene oxide, and 4 % of the weight of allyl glycidyl ether, and glass transition temperature was [ 1,250,000 and the amount of heat of fusions of -63 degrees C and weight average molecular weight ] 61 J/g.

[0076] 1g [ of this polyether copolymer ] and cross linking agent dicumyl peroxide 0.015g was dissolved in acetonitrile 20ml, and 5ml of tetrahydrofuran solutions containing LiTFSI 0.15g was mixed. After carrying out the cast of this mixed liquor on the mold made from polytetrafluoroethylene and drying, under nitrogen-gas-atmosphere mind, it heated for 30 minutes and 140 degrees C of bridge formation films were obtained. The property of a bridge formation film was measured by the same approach as an example 1. the conductivity of a solid electrolyte --  $5.2 \times 10^{-5}$  S/cm it was .

[0077] Ethylene oxide / triethylene glycol methyl glycidyl ether / allyl-glycidyl-ether = 47/47/6 used in the example of comparison 2 example 5 3 yuan copolymer of weight % (weight average molecular weight 1,050,000) 1 g and cross linking agent benzoyl oxide 0.015g Acetonitrile 20ml It dissolves and is LiTFSI 0.15g. 5ml of tetrahydrofuran solutions was mixed. After carrying out the cast of this mixed liquor on the mold made from polytetrafluoroethylene and drying, under nitrogen-gas-atmosphere mind, 3hr heating was carried out and 100 degrees C of bridge formation films were obtained. conductivity of a solid electrolyte  $7.1 \times 10^{-5}$  S/cm it was .

[0078] The lithium metallic foil was used as the giant-molecule solid electrolyte obtained in the example 3 as example 6 electrolyte, and a negative electrode, the cobalt acid lithium (LiCoO<sub>2</sub>) was used as positive active material, and the rechargeable battery was constituted. size of a solid polymer electrolyte 10mm x 10mm x 0.1mm it is . size of a lithium foil 10mm x 10mm x 0.1mm it is . After the cobalt acid lithium mixed the lithium carbonate and cobalt carbonate fine particles of the specified quantity At 900 degrees C It prepared by calcinating for 5 hours. Next, cobalt acid lithium which ground this and was obtained It is acetylene black to 85 weight sections. Copolymer obtained in 5 weight sections and the example 1 30MPa after adding 10 weight sections and the LiTFSI 5 weight section and mixing with a roll By the pressure 10mm x 10mm x 0.5mm Press forming was carried out and it considered as the positive electrode of a cell.

[0079] While putting the pressure of 1MPa to insert the giant-molecule solid electrolyte obtained in the example 3 with a lithium metallic foil and a positive electrode, and for an interface stick it The charge-and-discharge property of a cell was investigated at 25 degrees C. Early terminal voltage The discharge current in 3.8 V It is 0.1 mA/cm<sup>2</sup> and was able to charge by 0.1 mA/cm<sup>2</sup>. Since the cell of this example is easily producible to a thin thing, moreover, it becomes a mass lightweight cell.

[0080] Polyether copolymer obtained in the example 7 example 1 1g and cross linking agent dicumyl peroxide 0.015g Propylene carbonate solution of LiTFSI 0.15g 0.5ml It mixed. At the bottom of nitrogen-gas-atmosphere



mind, and 140 degrees C after carrying out the cast of this mixed liquor on the mold made from polytetrafluoroethylene It heated for 30 minutes and the gel bridge formation film was obtained. conductivity of the solid electrolyte in 15 degrees C  $1.1 \times 10^{-3}$  S/cm it was .

[0081] 1g of polyether copolymers obtained in the example 8 example 2, cross linking agent benzoyl oxide 0.015g Tetraethylene glycol wood ether solution containing LiTFSI 0.15g 0.5ml It mixed. After carrying out the cast of this mixed liquor on the mold made from polytetrafluoroethylene, under nitrogen-gas-atmosphere mind, 3hr heating was carried out and 100 degrees C of gel bridge formation films were obtained. Conductivity of the solid electrolyte in 20 degrees C It was  $8.7 \times 10^{-4}$  S/cm.

[0082] 1g of polyether copolymers obtained in the example 9 example 2, cross linking agent dicumyl peroxide Tetraethylene glycol wood ether solution containing 0.015g and LiTFSI 0.15g 0.5ml It mixed. At the bottom of nitrogen-gas-atmosphere mind, and 140 degrees C after carrying out the cast of this mixed liquor on the mold made from polytetrafluoroethylene It heated for 10 minutes and the gel bridge formation film was obtained. Conductivity of the solid electrolyte in 20 degrees C It was  $7.8 \times 10^{-4}$  S/cm.

[0083] The lithium metallic foil was used as the gel giant-molecule solid electrolyte obtained in the example 7 as example 10 electrolyte, and a negative electrode, the cobalt acid lithium ( $\text{LiCoO}_2$ ) was used as positive active material, and the rechargeable battery was constituted. Size of a solid polymer electrolyte It is 10mm x 10mm x 0.1mm. size of a lithium foil 10 mm x 10mm x 0.1mm it is . After the cobalt acid lithium mixed the lithium carbonate and cobalt carbonate fine particles of the specified quantity At 900 degrees C It prepared by calcinating for 5 hours. next, cobalt acid lithium which ground this and was obtained 85 weight sections -- receiving -- acetylene black Copolymer obtained in 5 weight sections and the example 1 10 weight sections -- and -- Pressure of 30MPa after adding the LiTFSI 5 weight section and mixing with a roll 10mm x 10mm x 0.5mm Press forming was carried out and it considered as the positive electrode of a cell.

[0084] While putting the pressure of 1MPa to insert the giant-molecule solid electrolyte obtained in the example 7 with a lithium metallic foil and a positive electrode, and for an interface stick it The charge-and-discharge property of a cell was investigated at 25 degrees C. The discharge current of terminal voltage 3.8V of the first stage is 0.1 mA/cm<sup>2</sup>, and was able to be charged by 0.1 mA/cm<sup>2</sup>. Since the cell of this example is easily producible to a thin thing, moreover, it becomes a mass lightweight cell.

[0085] The lithium metallic foil was used as the gel giant-molecule solid electrolyte obtained in the example 8 as example 11 electrolyte, and a negative electrode, the cobalt acid lithium ( $\text{LiCoO}_2$ ) was used as a positive electrode, and the rechargeable battery was constituted. The size of a solid polymer electrolyte is 10mm x 10mm x 0.1mm. size of a lithium foil 10mm x 10mm x 0.1mm it is . After the cobalt acid lithium mixed the lithium carbonate and cobalt carbonate fine particles of the specified quantity At 900 degrees C It prepared by calcinating for 5 hours. Next, cobalt acid lithium which ground this and was obtained Copolymer obtained in the acetylene black 5 weight section and the example 2 to 85 weight sections By the pressure of 30MPa after adding 10 weight sections and the  $\text{LiClO}_4$  5 weight section and mixing with a roll 10mm x 10mmx 0.5mm Press forming was carried out and it considered as the positive electrode of a cell. the giant-molecule solid electrolyte obtained in the example 8 is inserted with a lithium metallic foil and a positive electrode, and an interface sticks it -- as -- The charge-and-discharge property of a cell was investigated at 25 degrees C, putting the pressure of 1MPa. Early terminal voltage The discharge current of 3.8V It is 0.1 mA/cm<sup>2</sup> and was able to charge by 0.1 mA/cm<sup>2</sup>. Since the cell of this example is easily producible to a thin thing, moreover, it becomes a mass lightweight cell.

[0086]

[Effect of the Invention] The solid polymer electrolyte of this invention is excellent in workability, a moldability, a mechanical strength, flexibility, thermal resistance, etc., and the ion conductivity improves remarkably. Therefore, a solid state battery (especially rechargeable battery) is begun, and the application to the application, the antistatic agent further for rubber or plastic material, or antielectric ingredient to electronic equipment, such as a mass capacitor and a display device, for example, electrochromic display etc., is expected.

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[Translation done.]